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POPULATION IRRADIATION DOSE ASSESSMENT FOR ^{14}C EMISSIONS FROM NPP WITH RBMK-1000 AND EGP-2 REACTORS

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The ^{14}C emissions from NPP with RBMK-1000 and EGP-6 reactors as well as the yearly population irradiation dose due to these emissions are evaluated. A model is presented for calculating the population irradiation dose due to ^{14}C emissions assuming an equilibrium ^{14}C distribution between local food products and atmospheric air and absence of such an equilibrium for humans as a result of the consumption of imported food products. This model makes it possible to legitimately reduce the conservatism of the evaluation of the dose from standard NNP emissions as compared with the operating procedures and recommendations. The estimated population irradiation dose from ^{14}C emissions is as follows, μSv/yr: 2.3 Smolensk, 1 Leningrad, 1.3 Kursk, and 0.034 Bilibino NPP. Irrespective of the computational model the ^{14}C contribution in the total population irradiation dose due to emissions from NPP with RBMK-100 and EGP-6 reactors is significantly higher than 1%, so that it must be normalized and monitored.

^{14}C is a radioactive isotope of carbon with half-life 5730 yr, emitting β-particles with average energy 49.3 keV. The main reaction resulting in its formation is the interaction of neutrons and nitrogen $^{14}\mathrm{N}(n,p)^{14}\mathrm{C}$. Other reactions – $^{15}\mathrm{N}(n,a)^{14}\mathrm{C}$, $^{16}\mathrm{O}(p,3p)^{14}\mathrm{C}$, $^{17}\mathrm{O}(n,\alpha)^{14}\mathrm{C}$, and $^{13}\mathrm{C}(n,\gamma)^{14}\mathrm{C}$ – make a very small contribution because of their small interaction cross section and the low content of the nuclei of these isotopes in the natural mixture of elements. Native ^{14}C is formed when cosmic-ray neutrons interact with the Earth's atmosphere. Its total content in the atmosphere is equal to $(1.4–2.2)\cdot10^{17}$ Bq [1]. Technogenic ^{14}C present in the atmosphere is mainly due to nuclear weapons tests. The total activity of ^{14}C which entered the atmosphere as a result of nuclear weapons tests is estimated to be $(2.1–2.5)\cdot10^{17}$ Bq [1]. Currently, the source of technogenic ^{14}C is the activity of the nuclear industry enterprises. The total emissions of ^{14}C from NPP and spent nuclear fuel reprocessing enterprises is equal to $2.6\cdot10^{14}$ Bq/yr [1]. The yearly ^{14}C specific emissions per unit energy produced is specific to NPP with different types of reactors. In terms of this index the NPP form the following sequence: GCR>AGR>LWGR(RBMK)> BWR>PHWR>PWR(VVER) [2–4].

The RBMK reactor with boiling water under pressure as the coolant and graphite as the moderator is characterized by the presence of a large amount of nitrogen in the core and the large mass of carbon in the moderator. These features result in significant formation of ^{14}C [5]. In NPP with RBMK ^{14}C is formed in the following systems [3]: coolant (multiple forced circulation loop), system cooling of the safety-and-control system [SCS] channels, loops for nitrogen-helium purging of the reactor space and nitrogen purging of the metal-structures of the circum-reactor space, and moderator. According to estimates [6], the main contribution in ^{14}C emissions into the environment is due to the coolant and the loops for nitrogen-helium purging of the reactor space and nitrogen purging of the metal structures of the circum-reactor space. During normal operation

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of a power-generating unit with RBMK-1000, the main contribution in the formation of ^{14}C is due to the nuclear reaction $^{14}N(n, p)^{14}C$ owing to the large cross-section for the interaction of neutrons with ^{14}N atoms and the high content of nitrogen isotopes in the natural mixture [7]. ^{14}C egresses through the decay gas-holder, filter, and discharge pipes.

In Russia, in contrast to foreign countries, before 2017 systemic control and monitoring of atmospheric emissions of ^{14}C during standard operation of NPP were not conducted, Standards for the maximum admissible discharges were not established, and its contribution was neglected when estimating the yearly population irradiation dose [8, 9]. In accordance with Rostekhnadzor's current requirements, normalization and monitoring are necessary for all radionuclides which are present in the emissions and whose total contribution in the yearly effective irradiation dose of individuals in a critical group of the population is ≥99% [10]. Since ^{14}C is present in NPP emissions, its yearly ingress into atmospheric air and its contribution in the population irradiation dose must be determined.

IAEA's conservative approach is used in the procedures and recommendations for determining the norms for the maximum admissible emissions for calculating the population radiation dose from ^{14}C. This approach assumes that equilibrium is established in the ^{14}C distribution between the human organism and atmospheric air [11–14].

In the present article we present estimates of ^{14}C emissions from RBMK-1000 and EGP-6 as well as the yearly population irradiation dose formed by them. Both the conservative method of calculation [11–13] and an alternative method making it possible to legitimately reduce the conservatism of estimates, taking into account the local-product fraction of the population's food basket, are used.

Materials and measurement methods. Currently, there are 14 operating units with uranium-graphite reactors in four NPP: 10 units with RBM-1000 (Leningrad, Smolensk, and Kursk NPP) and four units with ERP-6 (Bilibino NPP). The objects of investigation were their ventilation systems. A mobile sampling stand was installed in the impulse line of the standard sampling system. A uniform laminar flow of a gas-air medium entered the system of the air intake setup in the ventilation tube. The gas-air mixture flowed along the impulse tube into the sampling stand. ^{14}C was sampled in the form of carbon dioxide by the bubbling method based on the absorption of carbon monoxide (IV) from the gas-air mixture by solution based on the exchange reaction of carbon monoxide (IV) with sodium hydroxide with formation of sodium carbonate. Due to the continuous pumping of carbon monoxide (IV) through the bubbler and prolonged sampling, the ^{14}C is concentrated in the bubbler solution.

The gas-air mixture was extracted from standard impulse tubes of the ventilation system. The gas-air mixture entered the sampling system through a filter with catching efficiency ≥99.95% (HEPA filter) for preliminary purification from aerosols and then directed into a bubbler with distilled water to precipitate tritium in the form of HTO. Next, the gas-air mixture passed through silica gel to remove moisture and then entered a cascade consisting of two bubblers, filled with a solution of sodium hydroxide with molar concentration 2.5 mol/dm^3, and a single bubbler filled with a barium hydroxide (II) solution with molar concentration 0.35 mol/dm^3 in order to record the saturation of the solution in the two preceding bubblers. An insoluble white sediment precipitates when carbon dioxide gas enters the bubbler with barium hydroxide (II). At the completion of the sampling process, the contents of the bubblers were transferred into a plastic vessel for shipment to an analytical laboratory.

The volumetric activity of ^{14}C in gaseous emissions was established from measurements of the activity of a counting sample prepared from a solution of a specimen taking into account the volume of the pumped gas-air mixture, catching factor, and volume of the solution in the bubbler. The activity in the counting sample was measured by the liquid scintillation method using a QUANTULUS 1220 (Finland) ultralow-background spectrometer. The activity of ^{14}C was calculated from measurements of the counting rate of the impulses from the counting sample, the detection efficiency of the ^{14}C β-radiation when a spectrometer, and the quenching parameter of the sample.

Methods of calculating the population irradiation dose. ^{14}C enters the atmosphere as carbon dioxide $^{14}CO_2$ gas, mixing with stable carbon and native ^{14}C. The stable carbon ^{12}C together with the radioactive isotope ^{14}C form a biological cycle, being the main chemical element of biological systems. The primary process whereby atmospheric ^{14}C enters the human food chain is incorporation into the biomass of plants by means of photosynthesis. The radioactive carbon passes from the plant biomass into the animal biomass and then along the food chains into the human organism.

Theoretically, technogenic and native ^{14}C rapidly arrive in equilibrium with stable carbon in all biological objects. The ratio of the content of radioactive and stable carbon in tissues becomes a constant quantity, equal to the ratio of the content of radioactive and stable carbon in air [14]. Under natural conditions, without the influence of the anthropogenic factor, the equilibrium amount of ^{14}C is equal to 0.233 Bq per 1 g of stable carbon [13]. The natural content of stable content in air is equal to 0.18 g/m^3, and the native equilibrium activity of ^{14}C in air is equal to 0.042 Bq/m^3. In the presence of anthropogenic ^{14}C in air in addition to native ^{14}C, its content in biological objects increases proportionately.

The currently operative methods of calculating the maximum admissible emissions presuppose that for humans, just as for other biological objects, an equilibrium specific activity of ^{14}C becomes established in tissues and atmospheric air. Then a conservative estimate of the expected yearly internal irradiation dose from ^{14}C present in atmospheric air in the form of carbon dioxide ^{14}CO$_2$ gas can be obtained using the simplified relation [13]

$$D = DCF\cdot C_{^{14}\mathrm{C,a}}/C_{^{12}\mathrm{C,a}}, \tag{1}$$

where $DCF = 5.6\cdot 10^{-5}$ (Sv/yr)/(Bq/g) is a conversion factor between the yearly internal irradiation dose from ^{14}C (Sv/yr) and its concentration in human tissues per 1 g of stable carbon; $C_{^{14}\mathrm{C,a}}$ is the computed volumetric activity of ^{14}C in atmospheric air from NPP emissions at the individual's domicile, Bq/m^3; $C_{^{12}\mathrm{C,a}} = 0.18$ g/m^3 is the concentration of stable carbon in air.

Most controversial is the problem of equilibrium content of ^{14}C being established between the human organism and atmospheric air. Most ^{14}C, just as stable carbon, enters the human organism with food. In addition, a significant fraction of the food people consume is imported; the locally-produced fraction for some foods does not exceed 10%. For this reason, it is appropriate to suppose that the equilibrium is established between atmospheric air and local plant and animal products, while atmospheric air and the human body may not be in equilibrium with one another. In this case, Eq. (1) will strongly overstate the ^{14}C contribution in the irradiation dose in people.

If ^{14}C equilibrium between atmospheric air and the human organism is not assumed, then the following relation can be used to calculate the yearly irradiation dose from ^{14}C:

$$D_{^{14}\mathrm{C}} = \varepsilon_{\mathrm{inh}} C_{^{14}\mathrm{C,a}} + \varepsilon_{\mathrm{f}} \sum\nolimits_i \alpha_i R_i C_{^{14}\mathrm{C,f},i}, \tag{2}$$

where $\varepsilon_{\mathrm{inh}}$ is the dose conversion factor on inhalation of ^{14}C (for an adult human on inhalation of ^{14}C in CO$_2$ form, $6.2\cdot 10^{-12}$ Sv/Bq [15]); U is the breathing intensity (for an adult human, $8.1\cdot 10^3$ m^3/yr [16]); $C_{^{14}\mathrm{C,a}}$ is the volumetric activity of ^{14}C in air at the location of the person's domicile, Bq/m^3; ε_{f} is the dose conversion factor for ^{14}C intake with food (for an adult human, $5.8\cdot 10^{-10}$ Sv/Bq [16]); α_i is the ith local product fraction in the population food ration; R_i is the yearly consumption of the ith product by the population, kg; $C_{^{14}\mathrm{C,f},i}$ is the specific activity of ^{14}C in the ith local product, Bq/kg.

The ^{14}C content in plant product assuming its equilibrium is established between ^{14}C in atmospheric air and the local plant product is calculated from the relation [17]

$$C^{\mathrm{p}}_{^{14}\mathrm{C,f},i} = f_{p,i} C_{14\mathrm{C,a}}/C_{12\mathrm{C,a}}, \tag{3}$$

where $f_{p,i}$ is the carbon fraction in the ith plant product, kg C/kg product; $C_{^{14}\mathrm{C,a}}$ is the volumetric activity of ^{14}C in air at the location of the production of the plant product, Bq/m^3; $C_{^{12}\mathrm{C,a}} = 1.8\cdot 10^{-4}$ kg/m^3 is the carbon mass in CO$_2$ form per unit volume of air. The carbon fraction in vegetables is equal to 0.059, root vegetables and potatoes 0.046, fruits 0.062, and cereal grains 0.39 [17].

The ^{14}C content in animal produce assuming equilibrium established between atmospheric air and local animal produce is calculated from the elation [17]

$$C_{^{14}\mathrm{C,f},i} = f_{ap,i} f_{\mathrm{con},i} C_{^{14}\mathrm{C,a}}/C_{^{12}\mathrm{C,a}}, \tag{4}$$

where $f_{ap,i}$ is the carbon fraction in the ith animal product, kg C/kg product: 0.065 milk, 0.2 meat [17]; f_{con} is the contaminated animal feed fraction (assumed to be 1 in the present work); $C_{^{14}\mathrm{C,a}}$ is the volumetric activity of ^{14}C in air at the production location of the animal product, Bq/m^3; $C_{^{12}\mathrm{C,a}} = 1.8\cdot 10^{-4}$ kg/m^3 is the carbon mass in the CO$_2$ form per unit volume of air.

The volumetric activity of ^{14}C in air was taken to be the same at the individual domicile, the production location of the local plant and animal product, i.e., farming by humans was taking place at the domicile location. The volumetric activity of ^{14}C in air was calculated by the method of [13] using the standard gaussian model for the transport of radioactive substances in the atmosphere taking into account the local characteristics of impurity dispersion for each NPP. The population irradiation dose was determined for the formal critical point of the location – a hypothetical point of maximum yearly dose outside the sanitary-protection zone of the NPP.

TABLE 1. Yearly Consumption of Food Products by the Rural Population (adults) near NPP with RBMK-1000 and EGP-6, kg [13]

Food product	NPP			
	Smolensk	Leningrad	Kursk	Bilibino
Milk	361.1	330.3	254.8	216.5
Meat	87.2	98.8	93.7	84.5
Bread	110.3	89.3	89.5	106.4
Potatoes	89.7	90.9	103.4	59.6
Vegetables	84.1	106	101.9	63.2
Fruits	83	83.7	72.9	51.6

TABEL 2. Local Food Product Fraction in the Food Ration of the Rural Population (adults) near NPP with RBMK-1000 and EGP-6 [13], %

Food product	NPP			
	Smolensk	Leningrad	Kursk	Bilibino
Milk	8.8	2.8	34.3	0
Meat	1.6	3.9	45.7	0.8
Potatoes	60.8	50.6	90.7	0.8
Vegetables	59.2	48.9	89.3	5
Fruits	69.6	46.7	93.6	0.8

The yearly consumption of food products by the population near NPP including the local product fraction was determined by the Federal Research Center for Nutrition, Biotechnology, and Food Safety in 2016 as part of work on the preparation of procedures and establishment of norms for the maximum admissible emissions of radioactive substances by NPP into atmospheric air [13] (Tables 1, 2). In the method of [13], the consumption factor for locally produced food products is not evaluated for bread, so that in the calculations this parameter was taken to be 0.5 for the Smolensk, Kursk, and Leningrad NPP and 0 for the Bilibino NPP.

Results and discussion. The measured volumetric activity of ^{14}C in the ventilation pipes of NPP with RBMK-1000 and EGP-6 according to data from radiation-technical inspection is presented in Table 3. Since each NPP possess several sources of ^{14}C emission, the following are shown in Table 3: zones – minimum and maximum ^{14}C volumetric activity and an estimate of the total yearly emissions from NPP, obtained using data on the number of sources of emissions, yearly rate of emission and measured volumetric activity of ^{14}C for each ventilation pipe of NPP. Conservative estimates obtained by formally combining the critical points of the location from the emissions from each source at NPP are presented. A critical point is taken to mean a point where the maximum volume metric activity of ^{14}C in air obtains outside the sanitary-protection zone of the NPP.

The yearly population irradiation dose from ^{14}C emissions from NPP was calculated by two methods (Table 4):

– assuming ^{14}C equilibrium between the human organism and atmospheric air – using Eq. (1);

– assuming ^{14}C equilibrium between local food products and atmospheric air but absence of such an equilibrium for the human organism – using Eqs. (2)–(4).

As one can see from Table 4, taking into account the local products consumed by the population strongly affects the yearly dose from ^{14}C emissions. The model supposing absence of ^{14}C equilibrium between the human organism and atmospheric air because of the significant imported food fraction gives an estimated yearly dose that is lower than the equilibrium model by a factor of 2.1 for the Kursk NPP, 2.9 Smolensk, 3.7 Leningrad, and 250 Bilibino.

The contribution of the dose from ^{14}C intake with food as compared with inhalation is almost 100% for Kursk, Smolensk, and Leningrad NPP and 95.8% for the Bilibino NPP.

TABLE 3. ^{14}C Emissions from RBMK-1000 and EGP-6 According to Data from Radiation-Technical Inspection, Computed Volumetric Activity of ^{14}C in Air at Critical Points of the Location near NPP

NPP	Reactor type	^{14}C volumetric activity in ventilation pipes, Bq/m^3		Discharge, TBq/yr	Volumetric activity in air, 10^{-2} Bq/m^3
		minimum	maximum		
Smolensk	RBMK-1000	339	449	12.1	2.15
Leningrad		24	138	2.89	1.19
Kursk		27	457	2.28	0.897
Bilibino	EGP-6	1380	3900	2.46	2.72

TABLE 4. Computed Yearly Population Irradiation Dose from ^{14}C Emissions from NPP with RBMK-1000 and EGP-6, μSv

NPP	Model			
	equilibrium, taking into account all the radiation paths	non-equilibrium		
		Inhalation intake	Food intake	Taking into account all irradiation pathways
Smolensk	6.69	0.001	2.28	2.28
Leningrad	3.7	0.0006	1.01	1.01
Kursk	2.79	0.0005	1.31	1.31
Bilibino	8.46	0.0014	0.0325	0.0339

TABLE 5. Contribution of ^{14}C to the Total Yearly Population Irradiation Dose from Emissions from NPP with RBMK-1000 and EGP-6 at a Critical Location

NPP	Yearly dose, μSv			Contribution of ^{14}C to the dose according to the model,%	
	Neglecting ^{14}C	Taking into account ^{14}C by model		equilibrium	nonequilibrium
		equilibrium	nonequilibrium		
Smolensk	1.13	7.82	3.41	85.6	67
Leningrad	3.23	6.93	4.24	53.4	23.8
Kursk	6.21	9	7.53	31	17.5
Bilibino	0.19	8.65	0.224	97.8	15.1

In [7], the effective yearly dose from native ^{14}C intake is estimated to be 9.7 μSv and the effective yearly dose from ^{14}C due to nuclear tests 1.9 μSv. Thus, using the non-equilibrium model for estimation the additional contribution of emissions in the irradiation from ^{14}C for the adult population living near NPP with RBMK-1000 does not exceed 20% and EGP-6 – 0.3%.

The ^{14}C contribution in the total yearly dose of emissions from NPP with RBMK-1000 and EGP-6 was evaluated using data on the yearly emission of the technogenic radionuclides (H^3, ^{60}Co, ^{131}I, ^{134}Cs, ^{137}Cs, radioactive inert gases) [18]. As we can see from Table 5, ^{14}C makes a significant contribution in the total yearly population irradiation dose from emissions from NPP with RBMK-1000 and EGP-6. For this reason, an adjustment of the method used to calculate the dose from ^{14}C taking into account data on the consumption of local agricultural products by the population makes it possible to legitimately reduce the conservatism in the estimation of the population irradiation dose from the standard discharges from NPP. The computed total (taking all radionuclides into account) yearly dose from standard emissions from NPP with RBMK-1000 will decrease by a factor of 2.3 for the Smolensk NPP, 1.6 for Leningrad, 1.2 for Kursk, and 39 for Bilibino.

The dose quota for the emissions from NPP with RBMK-1000 and EGP-6 reactors during normal operation was set at the level 200 μSv/yr [19]. The norms for the admissible emissions from NPP are calculated on the basis of the limits on the yearly radiation dose at a level of the minimum significant value 10 μSv/yr. Thus, in the adjustments to the model used to calculate the ^{14}C contribution the sum of the ratios of the yearly NPP emissions to the admissible values will decrease from 0.78 to 0.34 for the Smolensk NPP, 0.69 to 0.42 for Leningrad, 0.9 to 0.75 for Kursk, and 0.87 to 0.022 for Bilibino.

It should be noted that irrespective of the model used for the calculations the ^{14}C contribution in the total population irradiation dose from emissions from NPP with RBMK-1000 and EGP-6 is significantly higher than 1%. For this reason, norms must be established for the maximum admissible emissions of ^{14}C and the ^{14}C content in the emissions from NPP with RBMK-1000 and EGP-6 must be controlled.

It is recommended that in the procedures and recommendations for normalizing the atmospheric emissions of radioactive substances provisions be made for using the described non-linear model, together with reliable data on the local product fraction in food consumption by the population, to calculate the population irradiation dose from the standard emissions of ^{14}C by nuclear complex enterprises.

Conclusions. According to data from radiation-technical inspections, the atmospheric emission of ^{14}C from NPP with different reactors is equal to $2.28 \cdot 10^{12}$–$1.21 \cdot 10^{13}$ Bq/yr for RBMK-1000 and $2.46 \cdot 10^{12}$ Bq/yr for EGP-6. The model used to calculate the population irradiation dose, assuming the presence of ^{14}C equilibrium between local food products and atmospheric air but no such equilibrium for the human organism, makes it possible to legitimately reduce the conservatism in the evaluation of the dose from standard NPP emissions as compared with the operating procedures and recommendations. The assessment of the ^{14}C contribution in the yearly population irradiation dose confirms the need for normalization and control of this radionuclide in emissions from NPP with RBMK-1000 and EGP-6. The total yearly population irradiation dose near NPP with RBMK-1000 and EGP-6 taking into account the ^{14}C contribution does not exceed the minimum significant value 10 μSv/yr.

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